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The Effect of Varying Primary Emissions on the Concentrations of Inorganic Aerosols Predicted by the Enhanced UK Photochemical Trajectory Model

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ABSTRACT

An enhanced Photochemical Trajectory Model (PTM) has been used to simulate concentrations of secondary inorganic aerosol (for the purposes of this work, sulphate, nitrate, chloride and ammonium) in PM_{10} over a two-month period at a rural site in central southern England (Harwell). Judged against a base year of 2007, emissions of precursor gases, SO_2, NO_x and NH_3 have been varied over plausible ranges, occurring across the UK only, mainland Europe only, or the whole of Europe. The model is able to reproduce observed non-linearities and shows that abatement is less than proportional in all cases. Additionally, abatement of sulphur dioxide leads to increased nitrate concentrations. The combination of a weak response of nitrate to reductions in NO_x emissions, and the effect of sulphur dioxide reductions in increasing nitrate is consistent with the very small recent observed trends in nitrate concentrations over the UK. A scenario for 2020 in which emissions of SO_2, NO_x and NH_3 fall to 64%, 75% and 96% respectively of their 2007 baseline levels across the whole of Europe shows a reduction of 2 μg m^{-3} in secondary inorganic aerosol which is 13% below the baseline case for a two month period in 2007, due mostly to a fall in sulphate and ammonium. As this was a relatively high pollution period, it is estimated that over a full year, the reduction is more likely to be around 1 μg m^{-3}.

Keywords: Secondary inorganic aerosol, sulphate, nitrate, trajectory model

INTRODUCTION

The European Union Limit Values for PM_{10} and $PM_{2.5}$ and the exposure reduction target for $PM_{2.5}$ (Official Journal, 2008; Harrison et al., 2012a), together with the imperative to protect public health are strong drivers to reduce airborne concentrations of PM_{10} and $PM_{2.5}$. As industrial and automotive sources of primary pollutants have steadily reduced in recent years, so the relative importance of secondary components of airborne particulate matter has become more pronounced. Chemical analysis of $PM_{2.5}$ samples collected in Birmingham in 2004-2006 showed that secondary ammonium sulphate and ammonium nitrate contributed 24.0% and 21.2% respectively of measured $PM_{2.5}$ mass (Yin and Harrison, 2008). Additionally, there was an appreciable contribution of secondary organic aerosol (19.4% derived from data in Harrison and Yin (2008)). Yin and Harrison (2008) also showed that on high pollution days when PM_{10} exceeds the daily limit value of 50 µg m^{-3}, ammonium nitrate makes a disproportionately large contribution to the overall particle mass (average 45.8%). Similar influences of secondary components on PM_{10} and $PM_{2.5}$ are operative also in London (Harrison et al., 2012b).

In analysing exceedences of the 24 hour PM_{10} limit value in the southern United Kingdom, Charron et al. (2007) attributed a high importance to regional pollution events associated particularly with elevated concentrations of secondary inorganic aerosol. It is well recognised that a substantial proportion of such particulate matter arises from precursor emissions in the European mainland (Harrison et al., 2012a) and consequently it is important to understand the relative contributions which abatement of UK and continental European precursor emissions respectively can make to reducing airborne concentrations of secondary inorganic particles. An additional factor to take into account is that a careful analysis of the relationship of airborne concentrations of sulphate to those of sulphur dioxide suggests significant non-linearities (Jones and Harrison, 2011), i.e. a reduction in sulphur dioxide emissions is associated with a less than proportionate reduction in sulphate.

In order to understand the impacts of abatement policies, it is necessary to predict the influences of reductions of precursor emissions upon future secondary pollutant concentrations. This can be done most effectively through use of a chemistry-transport model, but the applicability of many such models is limited by their large computational expense whose implication is that only a small number of scenarios can be run, and consequently the full range of possible future emissions cannot readily be studied. A number of models have been used to predict concentrations of particulate matter components within the European atmosphere. These include Eulerian models such as LOTOS-EUROS (Schaap et al., 2008), CHIMERE (Bessagnet et al., 2009), REM-CALGRID model (RCG) (Beekmann et al., 2007), and the Unified EMEP model (Simpson et al. 2011). The unified EMEP model has been used for policy development in Europe (Aas et al., 2007) and to address regional scale impacts of NO_x and SO_2 emission reductions on PM mass concentrations. More recently, Megaritis et al. (2012) have used the PMCAM$_x$-2008 model to evaluate the impact of 50% precursor emissions reductions upon $PM_{2.5}$ concentrations in Europe.

The CityDelta project compared the ability of several models to predict the impact of emissions reductions upon concentrations in European cities (Cuvelier et al., 2007), specifically Berlin, Milan, Paris and Prague (Thunis et al., 2007). A subsequent study (Stern et al., 2008) examined the ability of five chemical transport models to reproduce PM_{10} episode conditions in central Europe. Model specific studies, such as those with CHIMERE, have sought to simulate particulate matter concentrations in specific parts of Europe, e.g. Portugal (Monteiro et al., 2007) and northern Italy (de Meij et al., 2009). Air quality models used for calculating aerosol species over the UK include the Community Multiscale Air Quality model (CMAQ) (Chemel et al., 2010) and the Hull Acid Rain Model (HARM), (Metcalfe et al., 2005). CMAQ over-predicted O_3 and under-predicted aerosol species with the exception of sulphate (Chemel et al., 2010). The HARM and ELMO models (Whyatt et al., 2007) underestimated sulphate, nitrate and ammonium by a large margin, and chloride massively. In the work of Redington and Derwent (2002), the NAME model slightly

under-predicted measured sulphate values (average negative bias across three sites was 0.33 μg m^{-3}) although the annual average values of nitrate compared well.

This paper is concerned with the application of a Lagrangian trajectory model to provide the results of multiple abatement scenarios upon concentrations of secondary organic aerosol.

THE MODEL

The numerical model used in this work is a heavily modified version of the UK Photochemical Trajectory Model (PTM). The PTM has been widely used by Derwent and his collaborators, especially in relation to the modelling of ozone (Derwent et al., 1996; Derwent et al., 2003; Walker et al., 2009; Baker, 2010) and of secondary organic aerosol (Johnson et al., 2006; Derwent et al., 2009; Abdalmogith et al., 2006). For its application to secondary inorganic aerosol, the model has been modified in our group in a number of important aspects. These are described in detail by Beddows et al. (2012) and may be summarised as follows:

- Enhancements to better describe the boundary layer meteorological processes affecting pollutant concentrations. In particular, data for temperature, relative humidity and mixing depths are derived from the HYSPLIT model and insolation is now a function of Julian Day and subject to attenuation by cloud.
- A more sophisticated treatment of in-cloud oxidation of sulphur dioxide has been incorporated.
- Concentrations of sea salt aerosol are estimated from the parameterisation of Gong et al. (2008) and used as inputs to both chloride and sodium for the aerosol thermodynamic model.
- The partition of reactive gases such as ammonia and nitric acid between vapour and the condensed phase is now described through the thermodynamic model, ISORROPIA II which describes the equilibrium processes within an internally mixed aerosol as a function of temperature and relative humidity (Fountoukis and Nenes, 2007).

120 The model set-up involves the calculation of mean concentrations resulting from back trajectories
121 over a period in 2007, terminating at the Harwell site in southern England. The model has been
122 extensively validated against measured data from Harwell and its performance tested against
123 standard metrics with satisfactory outcomes (Beddows et al., 2012). Concentrations correspond to
124 the PM_{10} size range.
125
126 Recent decades have seen substantial reductions in the emissions of SO_2 and NO_x (Vestreng et al.,
127 2007; AQEG, 2007) and the range of reductions in emissions modelled allows for a further
128 reduction of 50% in these gases. Reductions in NH_3 are less certain, and it has been predicted that in
129 certain areas (e.g. United States; Pye et al., 2009), there may be future increases in NH_3 emissions.
130 For this reason a wider range including both increases and reductions from the baseline was chosen
131 for the modelled NH_3 emissions than for the modelled NO_x and SO_2 emissions.
132
133 **METHOD**
134 The Photochemical Trajectory Model (PTM) was run over the period 19 March 2007 to 19 May
135 2007 with various assumed emissions of oxides of nitrogen (NO_x), sulphur dioxide (SO_2) and
136 ammonia (NH_3), to predict inorganic particulate matter concentrations at Harwell. This is the same
137 period over which model validation was conducted (Beddows et al., 2012). Emission values were
138 taken from the NAEI on a 10 x 10 km grid for the United Kingdom (UK), and from EMEP on a 50
139 x 50 km grid for the rest of Europe (EU) as described in Beddows et al. (2012), with changes in
140 emissions modelled for these areas both separately and together. Emissions from shipping in local
141 seas were abated in both the UK-only and EU-only scenarios. The variation of emissions was
142 considered over the range of 100% to 40% for NO_x 100% to 50% for SO_2, and 120% to 70% for
143 NH_3, in 10% steps, where 100% is the baseline (2007) rate of emission.
144

The 2976 half-hourly calculated concentrations predicted by the PTM for each emission scenario were used to calculate the mean concentration, over the two month period, of particulate nitrate, ammonium, sulphate and chloride. Chloride, although largely primary is affected by the processes determining concentrations of secondary sulphate and nitrate and was therefore included. The mean concentrations of the precursor gases HNO_3 and HCl were also calculated.

RESULTS AND DISCUSSION

For the purpose of this study, variations were made to the emissions of three precursor pollutants, i.e. ammonia, NO_x and sulphur dioxide. The model was then used to compute the resultant concentrations of ammonium, nitrate, sulphate and chloride (and coincidentally nitric acid and hydrochloric acid vapours) and the sum of the four particulate components, which for the purpose of this report is described as secondary inorganic aerosol (SIA).

$$SIA = \sum SO_4^{2-} + NO_3^- + Cl^- + NH_4^+$$

Figure 1 shows an example of the effect of abatement policies applied uniformly across the UK and the remainder of Europe. In the three-dimensional plot, one pollutant has been held constant; in this case ammonia emissions have been held constant at 100% of their 2007 value. The plot then shows the concentrations of the species indicated resulting from levels of sulphur dioxide emission between 50% and 100% of 2007 emissions and of abatement of NO_x such that emissions are between 40% and 100% of those in 2007. In Figure 1, it may be seen that nitrate concentrations, as expected, fall with the abatement of NO_x emissions, but that the fall is reduced by the abatement of sulphur dioxide. This confirms that the model is reflecting the atmospheric chemistry well. The reason for this effect is that the abatement of sulphur dioxide reduces the formation of sulphate (as seen in the graph for sulphate), which in turn reduces the irreversible uptake of ammonia into ammonium sulphate. This in turn implies that more ammonia remains in unneutralised form in the

atmosphere which then contributes to additional formation of nitrate by reaction with nitric acid. Abatement of sulphur dioxide and NO_x appears to contribute monotonically to a reduction in ammonium in the aerosol, but also leads to modestly increasing concentrations of chloride in the particles. This is a consequence of diminished scavenging of ammonia by sulphuric acid which is then free to react with hydrogen chloride, leading to a reduction in this species, seen in Figure 1, and an increase in particulate chloride. The overall effect of abatement of NO_x to 40% and SO_2 to 50% of 2007 emissions at constant ammonia emissions is a reduction of secondary inorganic aerosol to around 11 μg m^{-3} from an initial concentration slightly in excess of 15 μg m^{-3}. Figure S1 (Supplementary Information) shows the effect of reductions of emissions within the UK only. Taking the 100% ammonia scenario, secondary inorganic aerosol concentrations are reduced by a smaller margin from a little over 15 μg m^{-3} to around 12 μg m^{-3}. In Figure S2, the projections of emissions reduction for the European mainland, excluding the UK, show for a 100% ammonia scenario (i.e. 2007 emission levels for ammonia), a maximum reduction of secondary inorganic aerosol to around 13 μg m^{-3} from 15 μg m^{-3}.

Comparative charts of the concentrations of SIA, nitrate, ammonium, sulphate and chloride when emissions of NO_x, SO_2 and NH_3 from UK, EU and both areas together are separately reduced, while the other emissions are held at 2007 values (100%), are presented in Figures S3 to S7 (Supplementary Information) respectively. When NO_x or SO_2 emissions are reduced, the reduction of emissions from the United Kingdom (UK) has a greater effect on concentrations of sulphate, nitrate and chloride than the reduction of emissions from the rest of Europe (EU). In the case of ammonium, both appear equally effective. These results appear in tabulated form in Tables S1-3 (in Supplementary Information).

Figure S4 shows a considerable beneficial effect of NO_x abatement upon nitrate concentrations especially if that reduction is across both the UK and mainland Europe. However, the benefits are

far less than proportionate. Figure S4 also confirms an increase in nitrate with the abatement of sulphur dioxide as mooted in the earlier text. On the other hand, abatement of ammonia emissions has a decidedly beneficial effect upon nitrate concentrations. The effects of abatement policies for sulphur dioxide, NO_x and ammonia are all beneficial in reducing ammonium concentrations with the greatest benefits coming from reductions across the whole of Europe, including the UK. Reducing ammonia emissions appears to be the most effective measure when comparing the 100% to 70% scenarios, which is consistent with the results of Megaritis et al. (2012). Figure S7 shows that effects on chloride concentrations of reductions in NO_x, SO_2 and ammonia are relatively modest but not zero. When these changes are summed as secondary inorganic aerosol (Figure S3), it is seen that even quite major reductions in emissions of precursor compounds have only a relative modest effect upon the concentration of secondary inorganic aerosol.

The general trends in concentration with respect to NO_x and SO_2 emissions are consistent across all NH_3 emission levels, and all geographic areas for which emissions were varied. SIA, ammonium and HNO_3 concentrations reduce with reductions in both NO_x and SO_2 emissions. Nitrate falls with reducing NO_x emissions but rises with cuts in SO_2 emissions (Figure S4). Sulphate falls with reducing SO_2 and ammonia emissions but is largely unaffected by reduced NO_x emission (Figure S6). Chloride increases slightly with reductions in SO_2 and NO_x emissions (Figure S7). According to Figure S3, SIA is relatively insensitive to reductions in a single precursor. A much greater reduction in SIA is seen when all three precursors are reduced by the maximum amount.

Secondary Pollutant – Precursor Relationships

These appear in Figures 2 to 4. Figure 2 shows the relationship of atmospheric concentrations of sulphate to sulphur dioxide which portrays the non-linearity widely believed to affect this relationship, and which was inferred from measured atmospheric concentrations by Jones and Harrison (2011). This graph also demonstrates the relative ineffectiveness for UK sulphate

concentrations of reducing only emissions in mainland Europe, but also that the reduction across both the UK and mainland Europe is more effective than reductions in the UK alone. Figure 3 shows the corresponding relationship between nitrate and emissions of NO_x. This portrays a broadly similar pattern of behaviour but the apparent intercept in nitrate at zero NO_x derived from curve fitting seems implausible and there must be a steepening relationship as NO_x is reduced further. The relative shallowness of this curve over the abatement range tested appears consistent with the fact that nitrate concentrations in UK air appear relatively insensitive to NO_x emission reductions in recent years. For example, data from UK rural sites showed a marked decline in NO_2 between 1987 and 2007, but no trend in particulate nitrate between 1999 and 2007 (AEA, 2008). It also shows clearly the relative importance of UK emissions in influencing NO_x far more than nitrate concentrations which are quite strongly influenced by European mainland emissions. Figure 4 shows the corresponding relationship between ammonium and ammonia emissions. This also shows a non-linearity but a significant beneficial impact of emissions reductions. Figures 5 and S8 show the impacts of sulphur dioxide abatement upon nitrate and ammonium respectively. Figure 5 shows clearly that nitrate has a tendency to increase as sulphur dioxide is reduced as indicated in the discussion above. On the other hand, reductions in sulphur dioxide emissions have clear benefits in reducing ammonium concentrations (Figure S8) as do reductions in NO_x in reducing ammonium (Figure S9). Figure 6 shows that reductions of NO_x lead to increases in sulphate. The effect is relatively small but it is most marked for reductions in mainland Europe. This is presumably an effect of lesser competition for oxidising agents with a reduction in concentrations of NO_x leading to enhanced oxidation of sulphur dioxide. Substantial benefits arise from the abatement of ammonia. This is seen in respect of sulphate concentrations in Figure 7 and of nitrate concentrations in Figure S10, both of which benefit significantly from ammonia abatement, as do concentrations of ammonium (Figure 4).

Comparison with the Work of Derwent et al. (2009)

The paper by Derwent et al. (2009) is the only broadly similar study with which to compare our results. It is also based upon a moving air parcel trajectory model but averages over each day of 2006 at Harwell, Oxfordshire, rather than focussing on a more limited time period with a minor pollution episode as in this study. In comparison to this study, it investigates a more limited range of abatement scenarios and presents results relating only to 30% reductions in emissions of individual pollutants rather than the mixed abatement cases run in our study. The model of Derwent et al. (2009) uses a "kinetic" approach to simulating formation of aerosol constituents whereas this work uses kinetics to determine the formation of oxidised products and thermodynamics to simulate their incorporation into particles. One difficulty with the thermodynamic approach is selecting an appropriate temperature/humidity condition at which to simulate the particle-vapour equilibria, which can be very sensitive to these variables. Another issue for nitrate is the presence of artefacts (generally negative) in the measurements, which will affect both modelling approaches. Nonetheless, both models perform well in simulating the magnitude of current nitrate concentrations (Beddows et al., 2012; Derwent et al., 2009). Table S4 (Supplementary Information) shows the concentrations predicted by the Derwent et al. (2009) model in comparison to those in the current study and Table 1 shows percentage reductions in each case from the base case for the 30% reduction scenarios presented by Derwent and co-workers.

In the case of sulphur dioxide abatement, it may be seen from Table 1 that the general pattern of behaviour is the same, and within each model a sulphate reduction arising from sulphur dioxide abatement in the UK only and rest of Europe cases and a substantially greater abatement resulting from reductions across the board. The main difference is that Derwent et al. (2009) predicted a 70% sulphate concentration from across the board abatement, whereas in the current work, the predicted reduction in sulphate is to a level of 78%. The current work predicts 89% for both the UK only and rest of Europe reductions whereas Derwent et al. (2009) predicted 85%. Once again, a

broadly similar pattern of outcomes is seen for abatement of NO_x with greater reductions shown by Derwent et al. (2009) (an 80% nitrate level as compared to 87% in our model), with a greater effect of abatement in the rest of Europe (88% nitrate) in the Derwent et al. (2009) model compared to the UK only case (92% nitrate), comparing with 94% nitrate in both scenarios in our model. In common with our model, the Derwent et al. (2009) model shows an increase in nitrate with sulphur dioxide abatement, although the effect is less in the Derwent et al. (2009) model for across the board abatement but greater for rest of Europe abatement than in our model. In the case of ammonia abatement, Derwent et al. (2009) show a reduction in ammonium to 79% in the across the board case as opposed to 83% in our model. Both models show 90% ammonium in the UK only case and Derwent et al. (2009) show 89% ammonium in the Europe-only case, whereas in our model it is 94%. Both models show ammonia abatement to be beneficial for all measured species. Given the differences both in model formulation, and the scenarios run (annual means versus a two month episode in different years), the agreement is suprisingly close.

Predictions for the European 2020 Scenario

AQEG (2012) gives projections of UK precursor emissions in 2020, together with emission for 2005 and 2010 in respect of the EU-27 and shipping in European waters. Baseline EU-27 and shipping emissions were estimated by averaging the 2005 and 2010 emissions in the AQEG report. The 2020 emissions (sum of EU-27 and shipping) were estimated as a percentage of baseline as SO_2, 64%; NO_x, 75% and NH_3, 96%.

The results of this scenario appear in Table 2, which shows the all of Europe, mainland only and UK-only reduction. It shows 2020 concentrations of SIA of 2.0 µg m^{-3} below the baseline case, a reduction of 13%, due mostly to a fall in sulphate and ammonium. The mean PM_{10} concentration measured at Harwell over the period 19/3/2007 to 19/5/2007 was 28.2 µg m^{-3}, while that over the full year of 2007 was 21.5 µg m^{-3}. Mean concentrations of chloride, nitrate and sulphate over the 2-

month period modelled were 1.18, 6.64 and 3.89 μg m^{-3}, somewhat higher than that for the full year of 1.38, 3.06 and 2.40 μg m^{-3} respectively. Thus nitrate and sulphate were 46% and 62% respectively over the full year relative to the modelled period. Consequently, an overall reduction of about 1 μg m^{-3} in PM_{10} might be anticipated of over the full year as a result of the estimated 2020 emissions reductions relative to 2007.

CONCLUSIONS

These results demonstrate that the enhanced PTM is an effective way of investigating complex emission reduction scenarios. The results give a clear view of pollutant interactions and show reductions in some cases, and in others increases in concentrations that are qualitatively the same as Derwent et al. (2009) and quantitatively relatively similar. The differences are probably explained to a large degree by the lower concentrations in the base case in the Derwent et al. (2009) model and the non-linearities accounting for greater percentage reductions in that situation. However, the great benefit of the current work is that it investigates a much wider range of scenarios than those studied by Derwent et al. (2009), therefore allowing an assessment of likely future concentrations over a very wide range of plausible mitigation policies and scenarios. It also gives predictions for specific scenarios, such as that for 2020.

In the 2020 scenario, despite substantial reductions in both NO_x and SO_2 (by 25% and 36% respectively), the reduction in SIA is almost wholly due to a fall in the sulphate concentration. Nitrate, which already exceeds sulphate in the southern UK, and becomes dominant under episodic conditions (Yin and Harrison, 2008), responds little to the reduction in NO_x, reflecting the low sensitivity of nitrate to NO_x emission reductions seen in Figure S9 and the effect of decreasing SO_2 emissions in increasing nitrate (Figure 5). This is reflected in the very small changes in nitrate in the UK in recent years, despite substantial reduction in NO_x emissions.

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Derwent et al., 2009	**mean 15.00z values, 2006**				**Current work**	**19 March – 19 May 2007**			
With respect to base case values	**Sulphate**	**Nitrate**	**Ammonium**	**$PM_{2.5}$**	**With respect to no emission reduction**	**sulphate**	**nitrate**	**ammonium**	**SIA**
Across the board cases					**Reductions in all of Europe**				
30% SO_2 case	70%	105%	94%	93%	30% reduction in SO_2	78%	108%	87%	94%
30% NO_x case	105%	80%	92%	96%	30% reduction in NO_x	101%	87%	94%	95%
30% NH_3 case	100%	83%	79%	92%	30% reduction in NH_3	97%	86%	83%	91%
UK-only cases					**Reductions in UK only**				
30% SO_2 case	85%	101%	99%	97%	30% reduction in SO_2	89%	104%	94%	97%
30% NO_x case	103%	92%	97%	99%	30% reduction in NO_x	100%	94%	98%	97%
30% NH_3 case	100%	92%	90%	96%	30% reduction in NH_3	97%	92%	90%	94%
Rest of Europe-only cases					**Reductions in rest of Europe only**				
30% SO_2 case	85%	115%	96%	97%	30% reduction in SO_2	89%	104%	93%	97%
30% NO_x case	101%	88%	94%	97%	30% reduction in NO_x	101%	94%	97%	98%
30% NH_3 case	100%	91%	89%	96%	30% reduction in NH_3	100%	94%	94%	97%

Table 1: Comparison of Derwent et al. (2009) and present work – proportionate reductions

Species	No abatement	All area reductions		Mainland only reductions		UK only reduction	
	SO_2 = 100% NO_x = 100% NH_3 = 100%	**SO_2 = 64% NO_x = 75% NH_3 = 96%**		**SO_2 = 64% NO_x = 75% NH_3 = 96%**		**SO_2 = 64% NO_x = 75% NH_3 = 96%**	
	concentration	**concentration**	**difference**	**concentration**	**difference**	**concentration**	**difference**
Chloride	1.53	1.58	-0.05	1.56	-0.03	1.54	-0.01
Nitrate	5.98	5.85	0.13	5.97	0.01	5.93	0.05
Ammonium	1.80	1.40	0.41	1.61	0.20	1.62	0.19
Sulphate	5.75	4.24	1.50	5.04	0.70	4.96	0.79
SIA	15.06	13.07	1.99	14.18	0.88	14.04	1.02

Table 2: Estimated concentrations (μg m^{-3}) resulting from an assumed 2020 scenario (SO_2, 64%; NO_x, 75%; NH_3, 96%)

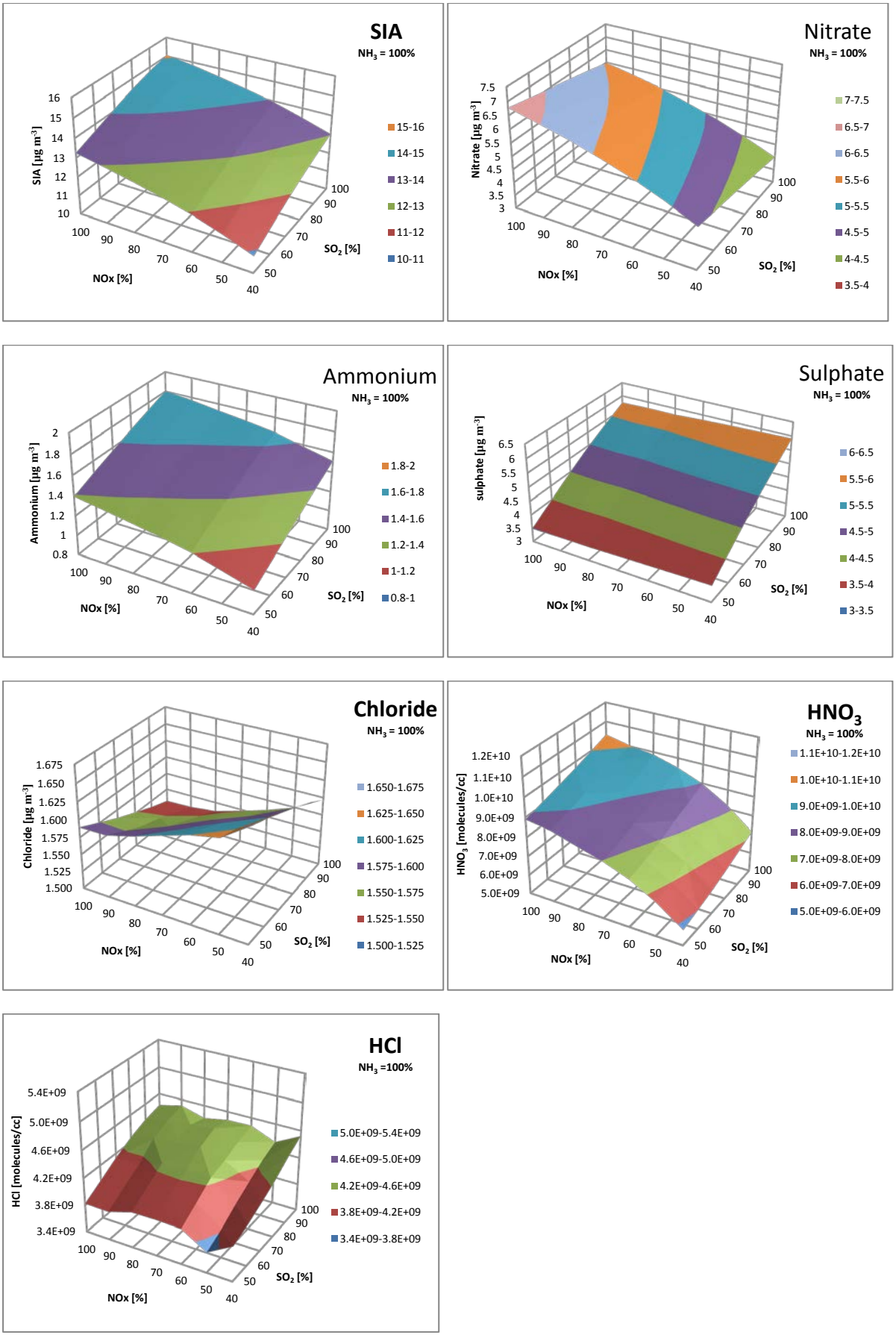

Figure 1: Effect of emission reductions in UK and Europe, for NH_3 = 100%

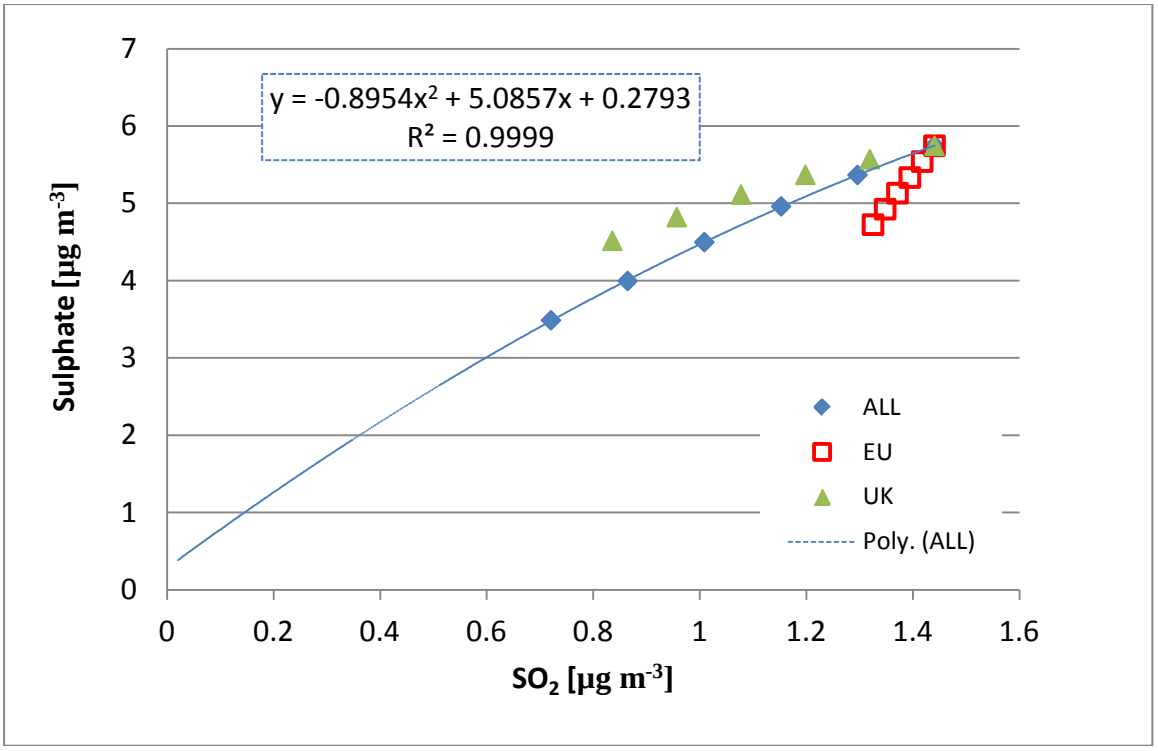

Figure 2: Predicted sulphate as a function of SO_2 at Harwell (SO_2 emissions varied in UK and Europe)

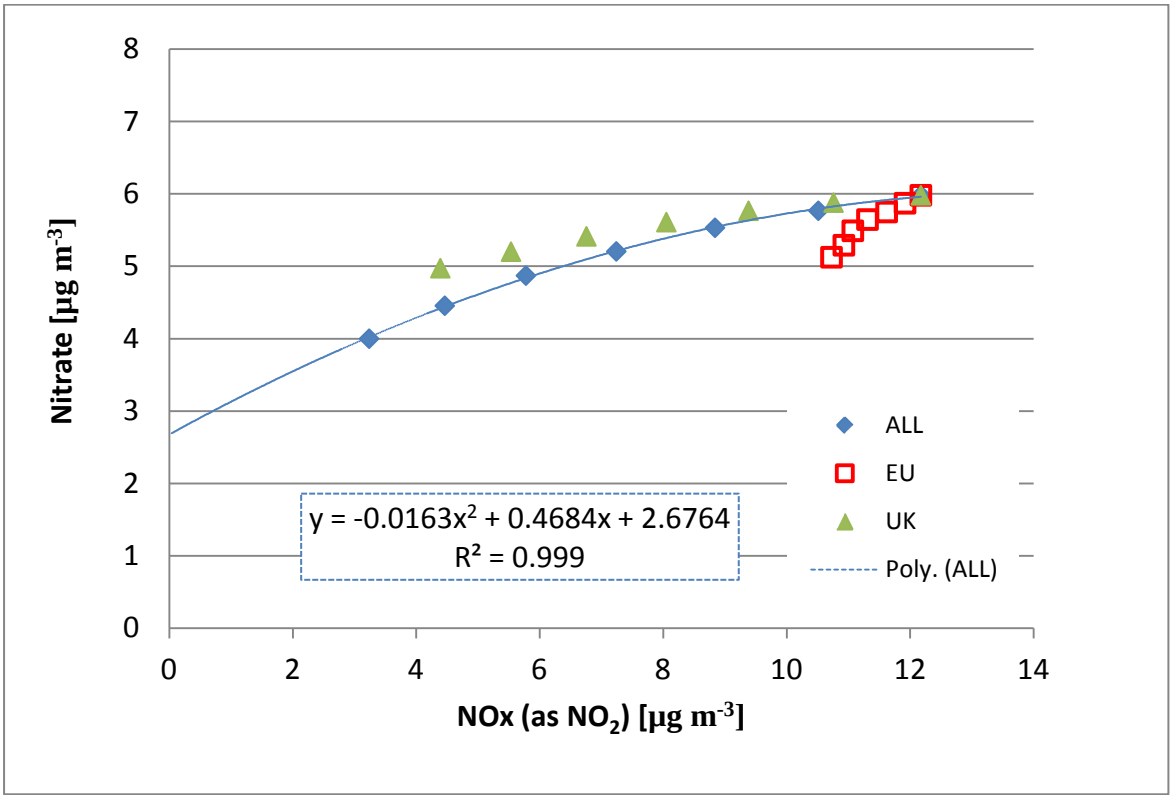

Figure 3: Predicted nitrate as a function of NO_x (as NO_2) at Harwell (NO_x emissions varied in UK and Europe)

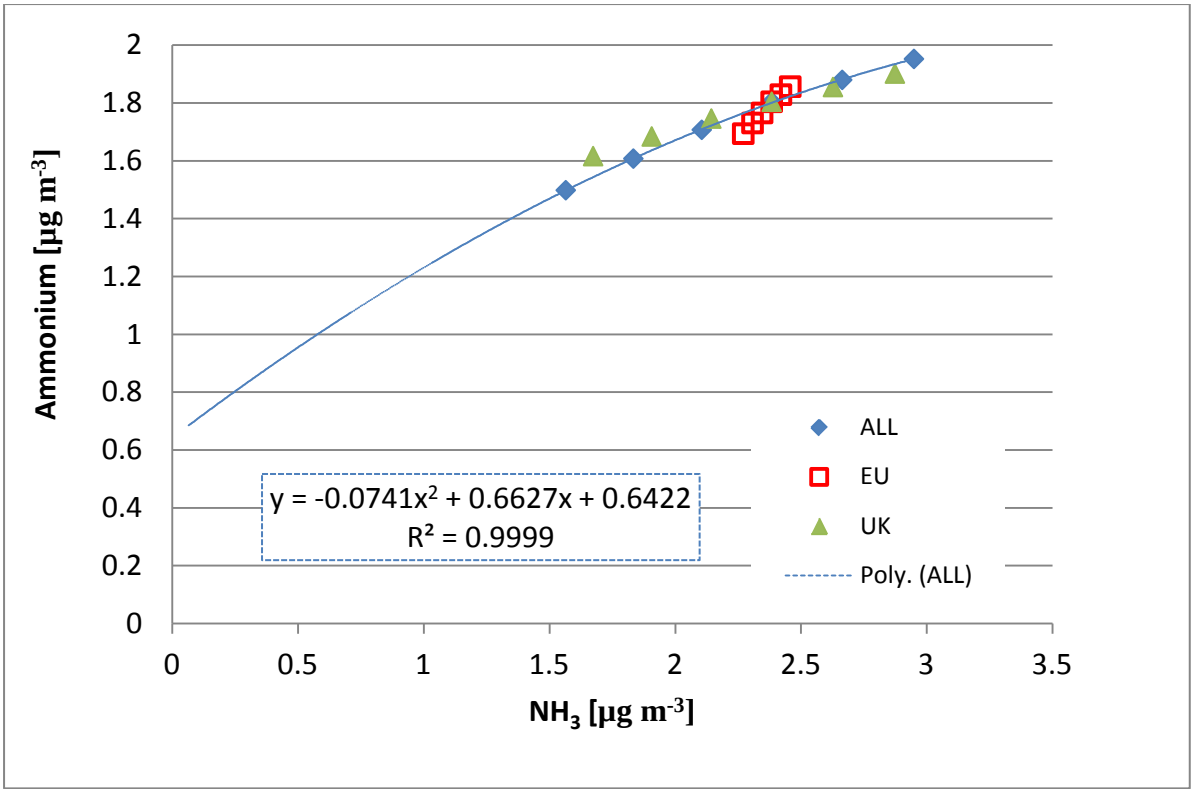

Figure 4: Predicted ammonium as a function of NH_3 at Harwell (NH_3 emissions varied in UK and Europe)

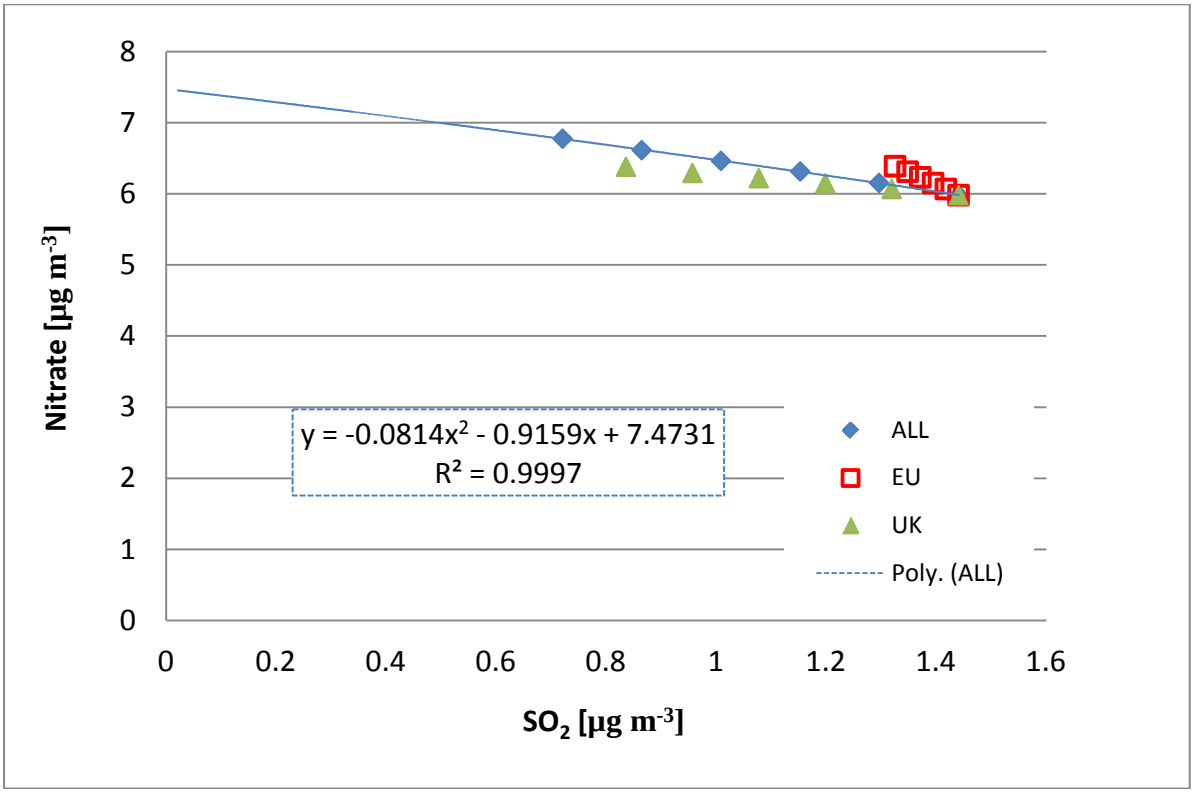

Figure 5: Predicted nitrate as a function of SO_2 at Harwell (SO_2 emissions varied in UK and Europe)

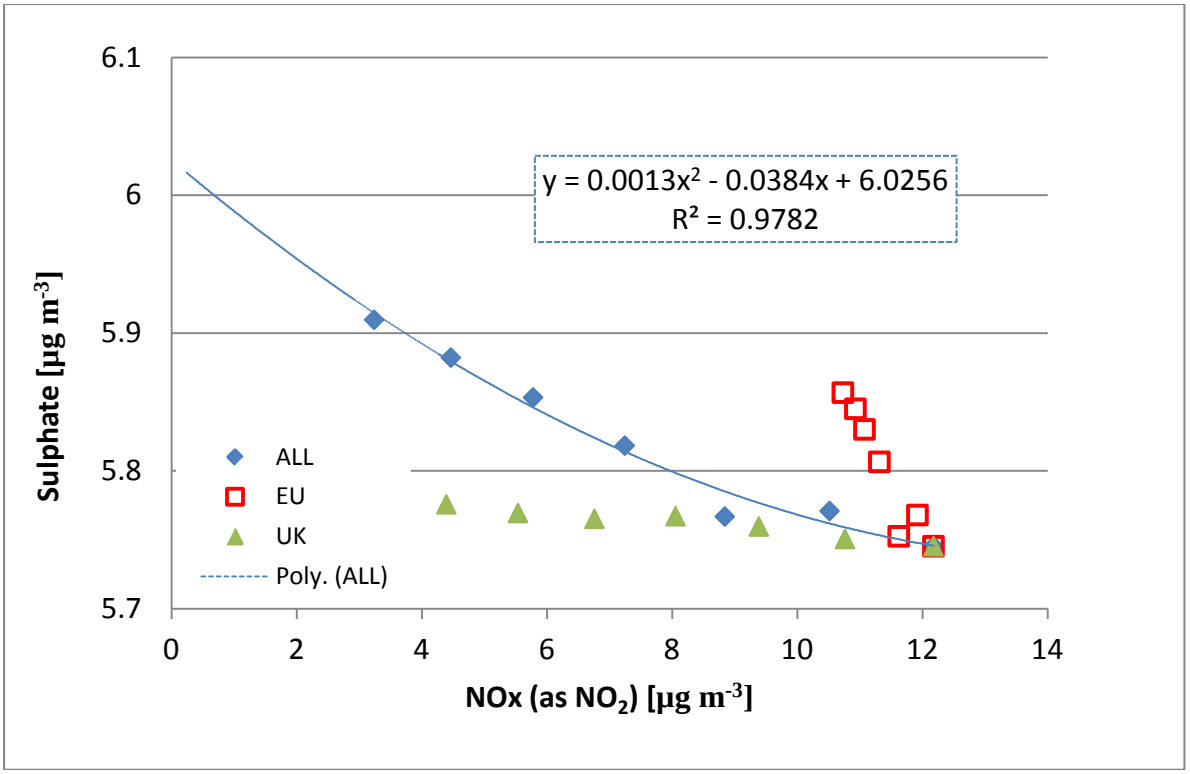

Figure 6: Predicted sulphate as a function of NO_x (as NO_2) at Harwell (NO_x emissions varied in UK and Europe)

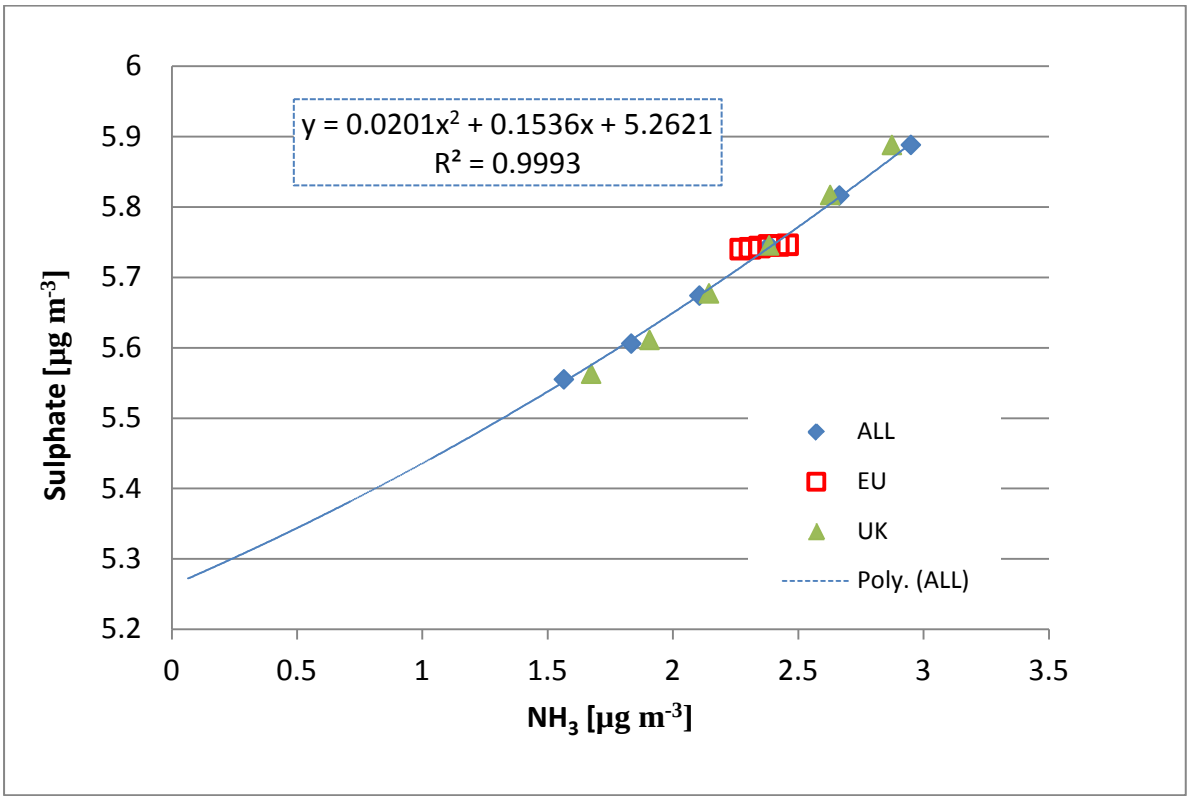

Figure 7: Predicted sulphate as a function of NH_3 at Harwell (NH_3 emissions varied in UK and Europe)